

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

To be Assigned

U.S. APPLICATION NO (if known, see 37 CFR 1.5

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

Mo-629/LEA33592
PRIORITY DATE CLAIMED

PCT/EP00/02242

14 March 2000 (14.03.00)

27 March 1999 (27.03.99)

TITLE OF INVENTION FLAME-RESISTANT POLYCARBONATE MOLDING MATERIALS MODIFIED WITH GRAFT POLYMERS

APPLICANT(S) FOR DO/EO/US 1) Thomas Eckel; 2) Michael Zobel; 3) Dieter Wittmann; 4) Torsten Derr; 5) Bernd Keller

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☒ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:

PTO Form 1449 and references listed therein; Preliminary Amendment w/Abstract

U.S. APPLICATION NO. 09/1937469
To be AssignedINTERNATIONAL APPLICATION NO
PCT/EP00/02242ATTORNEY'S DOCKET NUMBER
Mo-6621/LeA 33,59221. ☒ The following fees are submitted:**BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):**Neither international preliminary examination fee (37 CFR 1.482)
nor international search fee (37 CFR 1.445(a) (2)) paid to USPTO
and International Search Report not prepared by the EPO or JPO \$1000.00International preliminary examination fee (37 CFR 1.482) not paid to
USPTO but International Search Report prepared by the EPO or JPO \$860.00International preliminary examination fee (37 CFR 1.482) not paid to USPTO
but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00International preliminary examination fee (37 CFR 1.482) paid to USPTO
but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00International preliminary examination fee (37 CFR 1.482) paid to USPTO
and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00**ENTER APPROPRIATE BASIC FEE AMOUNT =****CALCULATIONS PTO USE ONLY**

\$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30
months from the earliest claimed priority date (37 CFR 1.492(e)).

\$

CLAIMS NUMBER FILED NUMBER EXTRA RATE

Total claims 18 -20 = 0 x \$18.00

Independent claims 1 -3 = 0 x \$80.00

MULTIPLE DEPENDENT CLAIM(S) (if applicable) + \$270.00

TOTAL OF ABOVE CALCULATIONS =

\$ 860.00

☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above
are reduced by 1/2.

\$ 0.00

SUBTOTAL =

\$ 860.00

Processing fee of \$130.00 for furnishing the English translation later than ☐ 20 ☐ 30
months from the earliest claimed priority date (37 CFR 1.492(f)).

\$

TOTAL NATIONAL FEE =

\$

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +

\$ 40.00

TOTAL FEES ENCLOSED =

\$ 900.00

Amount to be
refunded: \$

charged: \$

- a. ☐ A check in the amount of \$ _____ to cover the above fees is enclosed.
- b. ☒ Please charge my Deposit Account No. 13-3848 in the amount of \$ 900.00 to cover the above fees.
A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
overpayment to Deposit Account No. 13-3848. A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card
information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:



00157

PATENT TRADEMARK OFFICE

SIGNATUREJames R. Franks
NAME42,552
REGISTRATION NUMBER

09/937469

JCCB Rec'd PCT TO 25 SEP 2001

PATENT APPLICATION
Mo-6621
LeA 33,592

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICATION OF)
THOMAS ECKEL ET AL) PCT/EP00/02242
SERIAL NUMBER: TO BE ASSIGNED)
FILED: HERewith)
TITLE: FLAME-RESISTANT)
POLYCARBONATE MOLDING)
MATERIALS MODIFIED WITH)
GRAFT COPOLYMERS)

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents

Washington, D.C. 20231

Sir:


This preliminary amendment is being filed concurrently with the subject patent application. Upon granting a Serial Number and filing date, please amend the subject patent application as follows.

"Express Mail" mailing label number ET146899225US
Date of Deposit September 25, 2001

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Assistant Commissioner of Patents and Trademarks, Washington, D.C. 20231

Donna J. Veatch

(Name of person mailing paper or fee)


Signature of person mailing paper or fee)

Please amend the Application as follows.

IN THE ABSTRACT:

Please replace the abstract with the following.

--FLAME-RESISTANT POLYCARBONATE MOLDING MATERIALS MODIFIED
WITH GRAFT POLYMERS

ABSTRACT OF THE DISCLOSURE

Flame resistant graft polymer-modified polycarbonate molding compositions are described. The polycarbonate molding compositions comprise phosphorous compounds, and a coagulated mixture of fluorinated polyolefins or precompound fluorinated polyolefins. Polycarbonate molding compositions according the present invention are flame retardant, and have desirable mechanical properties, e.g., notched impact strength and elongation at break.--

A new abstract is included herewith on a separate sheet.

IN THE SPECIFICATION:

Please replace the title at line 1 of page 1 with the following.

--FLAME-RESISTANT POLYCARBONATE MOLDING MATERIALS MODIFIED
WITH GRAFT POLYMERS--

Please insert the following between lines 1 and 3 on page 1 of the specification.

--CROSS REFERENCE TO RELATED PATENT APPLICATIONS

The present patent application claims the right of priority under 35 U.S.C. 119 (a)-(d) and 35 U.S.C. 365 of International Application No. PCT/EP00/02242, filed 14 March 2000, which was published in German as International Patent Publication No. WO 00/58395 on 5 October 2000, which is entitled to the right of priority of German Patent Application No. 199 14 137.1, filed 27 March 1999.

FIELD OF THE INVENTION--

Please insert the following at line 8 on page 1 of the specification.

--BACKGROUND OF THE INVENTION--

Please insert the following at line 28 on page 2 of the specification.

--SUMMARY OF THE INVENTION--

Please insert the following at line 7 on page 4 of the specification.

--DETAILED DESCRIPTION OF THE INVENTION--

IN THE CLAIMS:

Please replace Claims 4, 6-13 and 15-18 with the following

4. (Once Amended, Clean) Moulding compositions according to Claim 1 containing 0.01 to 3 parts by weight of fluorinated polyolefin in the form of a coagulated mixture or as a precompound.

6. (Once Amended, Clean) Moulding compositions according to Claim 1, containing:

A) 40 to 99 parts by weight of aromatic polycarbonate and/or polyester carbonate;

B) 0.5 to 60 parts by weight of graft polymer prepared from,

B.1) 5 to 95 wt.% of one or more vinyl monomers, and

B.2) 95 to 5 wt.% of one or more grafting backbones having a glass transition temperature of $<10^{\circ}\text{C}$;

C) 0 to 45 parts by weight of at least one thermoplastic polymer selected from vinyl (co)polymers and polyalkylene terephthalates;

D) 0.5 to 20 parts by weight of said phosphorus compound represented

by formula (I); and

- E) 0.01 to 3 parts by weight of said fluorinated polyolefins in the form of said coagulated mixture or as said precompound.

7. (Once Amended, Clean) Moulding compositions according to Claim 1 wherein N in the formula (I) denotes an average value of 0.95 to 5.

8. (Once Amended, Clean) Moulding compositions according to Claim 1 wherein N in the formula (I) denotes an average value of 1 to 3.

9. (Once Amended, Clean) Moulding compositions according to Claim 6 containing fluorinated polyolefin or polyolefin mixture

- 1) as a coagulated mixture with at least one of components A to C, wherein the fluorinated polyolefin E or polyolefin mixture is mixed as an emulsion with at least one emulsion of components A to C and then coagulated,

or

- 2) as a precompound with at least one of components A to C, wherein the fluorinated polyolefins E are mixed as a powder with a powder or pellets of at least one of components A to C and melt-compounded.

10. (Once Amended, Clean) Moulding compositions according to Claim 1 containing one or more graft polymers prepared from,

B.1 5 to 95 wt.% of at least one vinyl monomer, and

B.2 95 to 5 wt.% of one or more grafting backbones having glass transition temperatures of $< 10^{\circ}\text{C}$.

11. (Once Amended, Clean) Moulding compositions according to Claim 10, containing as the vinyl monomers B.1 mixtures of,

B.1.1 50 to 99 parts by weight of at least one of vinyl aromatics, ring-substituted vinyl aromatics, and (meth)acrylic acid (C₁-C₈)-alkyl esters,

and

B.1.2 1 to 50 parts by weight of at least one of vinyl cyanides, (meth)acrylic acid (C₁-C₈)-alkyl esters, and derivatives of unsaturated carboxylic acids.

12. (Once Amended, Clean) Moulding compositions according to Claim 6 containing a diene rubber, acrylate rubber, silicone rubber or ethylene/propylene/diene rubber or mixtures thereof as the grafting backbone B.2.

13. (Once Amended, Clean) Moulding compositions according to Claim 1 wherein Y in the formula (I) denotes isopropylidene or methylene.

15. (Once Amended, Clean) Moulding compositions according to Claim 1 further containing vinyl (co)polymers of at least one monomer selected from vinyl aromatics, vinyl cyanides, (meth)acrylic acid (C₁-C₈)-alkyl esters, unsaturated carboxylic acids, and derivatives of unsaturated carboxylic acids.

16. (Once Amended, Clean) Moulding compositions according to Claim 1 further containing at least one additive selected from stabilisers, pigments, mould release agents, flow auxiliaries, antistatic agents, fillers and reinforcing materials.

17. (Once Amended, Clean) A method of using the moulding compositions according to Claim 1 for the production of mouldings.

18. (Once Amended, Clean) Mouldings prepared from moulding compositions according to Claim 1.

Mo-6621

REMARKS

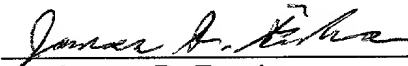
Claims in the case are 1-18. Claims 4, 6-13 and 15-18 have been amended herein. Claims 4, 6-13 and 15-18 of the above-identified patent application have been amended as to form, for example, by introducing indentation and replacing multiple dependencies with single dependencies.

The specification has been amended to include section headings, in accordance with accepted practice before the Office. The title has been changed to correspond to that of the related International Patent Publication No. WO 00/58395.

Page 1 of the application has been amended herein to introduce cross reference information. The cross reference information is presented in accordance with 37 C.F.R. 1.78(a)(2) (Federal Register / Vol. 65, No. 183 / Wednesday, September 20, 2000; Changes to Implement Eighteen-Month Publication of Patent Applications; Final Rule).

The amendments presented herein are not believed to represent the entry of new matter into the application. Applicants respectfully request entry of this preliminary amendment.

Respectfully submitted,

By 
James R. Franks
Agent for Applicants
Reg. No. 42,552

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VERSIONS WITH MARKINGS TO SHOW CHANGES MADE

IN THE ABSTRACT: (Marked-Up)

The abstract has been replaced with the following.

**FLAME-RESISTANT POLYCARBONATE MOLDING MATERIALS MODIFIED WITH
GRAFT POLYMERS**

ABSTRACT OF THE DISCLOSURE

Flame resistant graft polymer-modified polycarbonate molding compositions are described. The polycarbonate molding compositions comprise phosphorous compounds, and a coagulated mixture of fluorinated polyolefins or precompound fluorinated polyolefins. Polycarbonate molding compositions according the present invention are flame retardant, and have desirable mechanical properties, e.g., notched impact strength and elongation at break.

IN THE SPECIFICATION: (Marked-Up)

The following are changes and additions made to the specification.

The following is a version of the title of the application on line 1 of page 1, showing changes made thereto herein.

[Flame resistant, graft polymer-modified polycarbonate moulding compositions]
**FLAME-RESISTANT POLYCARBONATE MOLDING MATERIALS MODIFIED WITH
GRAFT POLYMERS**

The following has been inserted between lines 1 and 3 on page 1 of the specification.

CROSS REFERENCE TO RELATED PATENT APPLICATIONS

The present patent application claims the right of priority under 35 U.S.C. 119 (a)-(d) and 35 U.S.C. 365 of International Application No. PCT/EP00/02242, filed 14 March 2000, which was published in German as International Patent Publication No. WO 00/58395 on 5 October 2000, which is entitled to the right of priority of German Patent Application No. 199 14 137.1, filed 27 March 1999.

FIELD OF THE INVENTION

The following has been inserted at line 8 on page 1 of the specification.

BACKGROUND OF THE INVENTION

The following has been inserted at line 28 on page 2 of the specification.

SUMMARY OF THE INVENTION

The following has been inserted at line 7 on page 4 of the specification.

DETAILED DESCRIPTION OF THE INVENTION

IN THE CLAIMS: (Marked-Up)

The following are versions of the amended claims with markings to show changes made thereto in the present Preliminary Amendment.

4. (Once Amended, Marked-Up) Moulding compositions according to [claims 1 to 3,] Claim 1 containing 0.01 to 3 parts by weight of fluorinated polyolefin in the form of a coagulated mixture or as a precompound.

6. (Once Amended, Marked-Up) Moulding compositions according to [c]Claim 1, containing:

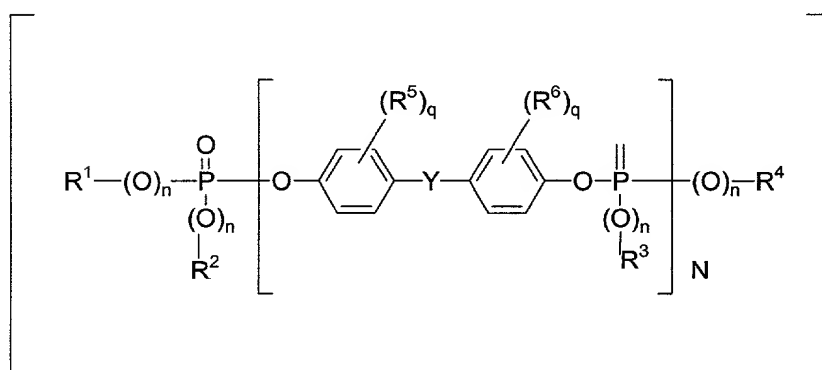
A) 40 to 99 parts by weight of aromatic polycarbonate and/or polyester carbonate;

B) 0.5 to 60 parts by weight of graft polymer [of] prepared from.

B.1) 5 to 95 wt.% of one or more vinyl monomers, [on] and

B.2) 95 to 5 wt.% of one or more grafting backbones having a glass transition temperature of <10°C[.];

- C) 0 to 45 parts by weight of at least one thermoplastic polymer selected from [the group comprising] vinyl (co)polymers and polyalkylene terephthalates[.];
- D) 0.5 to 20 parts by weight of [a] said phosphorus compound [of the] represented by formula (I); and



[in which R¹ to R⁶, Y, n, N and q have the meaning stated in claim 1,]

- E) 0.01 to 3 parts by weight of said fluorinated polyolefins [according to claim 1] in the form of said coagulated mixture or as said precompound.

7. (Once Amended, Marked-Up) Moulding compositions according to [one of the preceding claims,] Claim 1 wherein N in the formula (I) denotes an average value of 0.95 to 5.

8. (Once Amended, Marked-Up) Moulding compositions according to [one of the preceding claims,] Claim 1 wherein N in the formula (I) denotes an average value of 1 to 3.

9. (Once Amended, Marked-Up) Moulding compositions according to [one of the preceding claims,] Claim 6 containing fluorinated polyolefin or polyolefin mixture

- 1) as a coagulated mixture with at least one of components A to C, wherein the fluorinated polyolefin E or polyolefin mixture is mixed as an emulsion with at least one emulsion of components A to C and then coagulated,

or

- 2) as a precompound with at least one of components A to C, wherein the fluorinated polyolefins E are mixed as a powder with a powder or pellets of at least one of components A to C and melt-compounded.

10. (Once Amended, Marked-Up) Moulding compositions according to [one of the preceding claims] Claim 1 containing one or more graft polymers [of] prepared from,

B.1 5 to 95 wt.% of at least one vinyl monomer₁ [on] and

B.2 95 to 5 wt.% of one or more grafting backbones having glass transition temperatures of < 10°C.

11. (Once Amended, Marked-Up) Moulding compositions according to [c]Claim 10, containing as the vinyl monomers B.1 mixtures of₁

B.1.1 50 to 99 parts by weight of at least one of vinyl aromatics₁ [and/or] ring-substituted vinyl aromatics₁ and[/or] (meth)acrylic acid (C₁-C₈)-alkyl esters₁ and

B.1.2 1 to 50 parts by weight of at least one of vinyl cyanides₁ [and/or] (meth)acrylic acid (C₁-C₈)-alkyl esters₁ and[/or] derivatives of

unsaturated carboxylic acids.

12. (Once Amended, Marked-Up) Moulding compositions according to [one of the preceding claims,] Claim 6 containing a diene rubber, acrylate rubber, silicone rubber or ethylene/propylene/diene rubber or mixtures thereof as the grafting backbone B.2.

13. (Once Amended, Marked-Up) Moulding compositions according to [one of the preceding claims,] Claim 1 wherein Y in the formula (I) denotes isopropylidene or methylene.

15. (Once Amended, Marked-Up) Moulding compositions according to [one of the preceding claims,] Claim 1 further containing vinyl (co)polymers of at least one monomer selected from [the group comprising] vinyl aromatics, vinyl cyanides, (meth)acrylic acid (C₁-C₈)-alkyl esters, unsaturated carboxylic acids, [as well as] and derivatives [(such as anhydrides and imides)] of unsaturated carboxylic acids.

16. (Once Amended, Marked-Up) Moulding compositions according to [one of the preceding claims,] Claim 1 further containing at least one [addition] additive selected from [the group comprising] stabilisers, pigments, mould release agents, flow auxiliaries, [and/or] antistatic agents, fillers and reinforcing materials.

17. (Once Amended, Marked-Up) A method of using [Use of] the moulding compositions according to [one of the preceding claims] Claim 1 for the production of mouldings.

18. (Once Amended, Marked-Up) Mouldings [obtainable] prepared from moulding compositions according to [one of the preceding claims] Claim 1.

09/937469

Le A 33,592

-37- JC03 Rec'd PCT/PTO 25 SEP 2001

FLAME-RESISTANT POLYCARBONATE MOLDING MATERIALS
MODIFIED WITH GRAFT POLYMERS

ABSTRACT OF THE DISCLOSURE

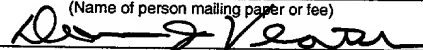
Flame resistant graft polymer-modified polycarbonate molding compositions are described. The polycarbonate molding compositions comprise phosphorous compounds, and a coagulated mixture of fluorinated polyolefins or precompound fluorinated polyolefins. Polycarbonate molding compositions according the present invention are flame retardant, and have desirable mechanical properties, e.g., notched impact strength and elongation at break.

09/937469

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Assistant Commissioner of Patents and Trademarks, Washington, D.C. 20231

Donna J. Veatch

(Name of person mailing paper or fee)



Signature of person mailing paper or fee)

Flame resistant, graft polymer-modified polycarbonate moulding compositions

JC03 Rec'd PCT/PTO

09/937469
25 SEP 2001

5 The present invention relates to flame resistant graft polymer-modified polycarbonate moulding compositions containing phosphorus compounds and specially prepared fluorinated polyolefins, which compositions exhibit excellent flame retardancy and very good mechanical properties such as notched impact strength, elongation at break and very good stress cracking behaviour.

10 Diphosphates are known as flame retardant additives. JP 59 202 240 describes the production of such a product from phosphorus oxychloride, diphenols such as hydroquinone or bisphenol A and monophenols such as phenol or cresol. These diphosphates may be used as flame retardants in polyamide or polycarbonate. However, this document makes no reference to improved stress cracking resistance due to the addition of the oligomeric phosphate to polycarbonate moulding compositions.

15 EP-A 363 608 describes flame resistant polymer blends prepared from aromatic polycarbonate, copolymer or graft copolymer containing styrene together with oligomeric phosphates as flame retardant. It is mentioned in general terms that tetrafluoroethylene polymers may be added.

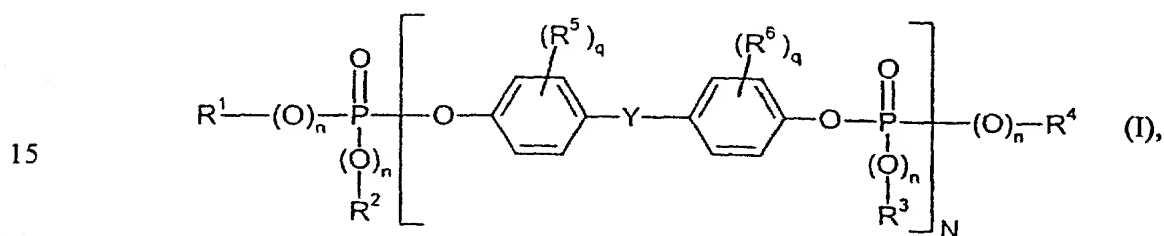
20 EP-A 0 767 204 describes flame resistant polyphenylene oxide (PPO) or polycarbonate mixtures which contain a mixture of oligophosphates (of the bisphenol A (BPA) oligophosphate type) and monophosphates as the flame retardant. Elevated flame retardant contents give rise to disadvantageous mechanical properties (notched impact strength, stress cracking behaviour) and reduced heat resistance.

25 EP-A 0 611 798 and WO 96/27600 describe moulding compositions which, in addition to polycarbonate, contain oligomeric, terminally alkylated phosphoric acid esters of the BPA type. Due to the alkylation, elevated contents are required in order

behaviour combined with elevated heat resistance. These moulding compositions are thus in particular suitable for those applications in which contact with special media, such as for example solvents, lubricants, cleaning agents, may occur.

5 It has now been found that graft polymer-modified polycarbonate moulding compositions which contain phosphorus compounds and special preparations of fluorinated polyolefins exhibit the desired range of properties.

10 The present invention accordingly provides graft polymer-modified polycarbonate moulding compositions containing phosphorus compounds of the formula (I)



in which

20 R^1 , R^2 , R^3 and R^4 are mutually independently C_1 - C_8 alkyl, optionally substituted by halogen, C_5 - C_6 cycloalkyl, C_6 - C_{10} aryl or C_7 - C_{12} aralkyl, each optionally substituted by halogen and/or alkyl,

25 n are mutually independently 0 or 1,

q are mutually independently 0, 1, 2, 3 or 4,

N is 0.9 to 10, preferably 0.95 to 5, in particular 1 to 3,

30 R^5 and R^6 mutually independently mean C_1 - C_4 alkyl, preferably methyl, or halogen, preferably chlorine and/or bromine,

Y means C₁-C₇ alkylidene, C₁-C₇ alkylene, C₅-C₁₂ cycloalkylene, C₅-C₁₂ cycloalkylidene, -O-, -S-, -SO-, -SO₂- or -CO-,

5 and fluorinated polyolefins in the form of a coagulated mixture or as a precompound.

The fluorinated polyolefins are preferably used as a coagulated mixture with at least one component selected from among polycarbonate, graft polymer and (co)polymer,
10 wherein the fluorinated polyolefin or polyolefin mixture is mixed as an emulsion with at least one emulsion of these components and then coagulated, or as a precompound with at least one of the stated components, wherein the fluorinated polyolefins or polyolefin mixture is mixed as a powder with a powder or pellets of at least one of these components and melt-compounded.

15 The thermoplastic moulding compositions preferably contain 0.5 to 20, particularly preferably 1 to 18 and in particular 2 to 16 parts by weight of phosphorus compound (I) or a mixture of phosphate compounds (I).

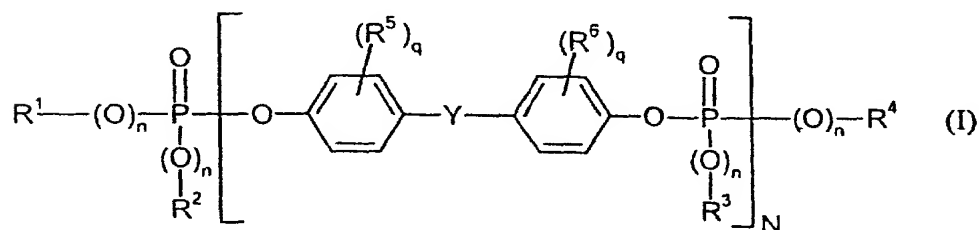
20 The thermoplastic moulding compositions preferably contain 0.01 to 3, in particular 0.05 to 2, very particularly preferably 0.1 to 0.8 parts by weight of fluorinated polyolefins in the form of special preparations, as a coagulated mixture or precompound.

25 Preferred thermoplastic moulding compositions are those containing

A) 40 to 99, preferably 60 to 98.5 parts by weight of aromatic polycarbonate and/or polyester carbonate

30 B) 0.5 to 60, preferably 1 to 40, in particular 2 to 25 parts by weight of graft polymer of

- B.1) 5 to 95, preferably 30 to 80 wt.% of one or more vinyl monomers on
- B.2) 95 to 5, preferably 20 to 70 wt.% of one or more grafting backbones having a glass transition temperature of $<10^{\circ}\text{C}$, preferably of $<0^{\circ}\text{C}$, particularly preferably of $<-20^{\circ}\text{C}$,
- C) 0 to 45, preferably 0 to 30, particularly preferably 2 to 25 parts by weight of at least one thermoplastic polymer selected from the group comprising vinyl (co)polymers and polyalkylene terephthalates,
- D) 0.5 to 20 parts by weight, preferably 1 to 18 parts by weight, particularly preferably 2 to 16 parts by weight of a phosphorus compound of the formula (I)



in which R^1 to R^6 , Y, n, N and q have the above-stated meaning,

- E) 0.01 to 3, preferably 0.05 to 2, particularly preferably 0.1 to 0.8 parts by weight of fluorinated polyolefins as
- E.1) a coagulated mixture with at least one of components A to C, wherein the fluorinated polyolefin or polyolefin mixture E is mixed as an emulsion with at least one emulsion of components A to C and then coagulated

or

- 5 E.2) as a precompound with at least one of components A to C, wherein the fluorinated polyolefin or polyolefin mixture E is mixed as a powder with a powder or pellets of at least one of components A to C and melt-compounded.

Component A

10 Component A aromatic polycarbonates and/or aromatic polyester carbonates which are suitable according to the invention are known from the literature or may be produced using processes known from the literature (*c.f.* in relation to the production of aromatic polycarbonates, for example Schnell, *Chemistry & Physics of Polycarbonates*, Interscience Publishers, 1964 and DE-AS 1 495 626, DE-OS 15 2 232 877, DE-OS 2 703 376, DE-OS 2 714 544, DE-OS 3 000 610, DE-OS 3 832 396; in relation to the production of aromatic polyester carbonates for example DE-OS 3 077 934).

20 Aromatic polycarbonates are produced for example by reacting diphenols with carbonic acid halides, preferably phosgene, and/or with aromatic dicarboxylic acid dihalides, preferably benzenedicarboxylic acid dihalides, by the phase interface process, optionally using chain terminators, for example monophenols, and optionally using trifunctional or greater than trifunctional branching agents, for example triphenols or tetraphenols.

25 Diphenols for the production of the aromatic polycarbonates and/or aromatic polyester carbonates are preferably those of the formula (III)

30



wherein

10

15



B

x

30

p

R^7 and R^8 mutually independently, individually selectably for each X^1 , mean hydrogen or C_1 - C_6 alkyl, preferably hydrogen, methyl or ethyl,

5 X^1 means carbon and

m means an integer from 4 to 7, preferably 4 or 5, providing that R^7 and R^8 are simultaneously alkyl on at least one atom X^1 .

10 Preferred diphenols are hydroquinone, resorcinol, dihydroxydiphenols, bis-(hydroxyphenyl)- C_1 - C_5 -alkanes, bis-(hydroxyphenyl)- C_5 - C_6 -cycloalkanes, bis-(hydroxyphenyl) ethers, bis-(hydroxyphenyl) sulfoxides, bis-(hydroxyphenyl) ketones, bis-(hydroxyphenyl) sulfones and α,α -bis-(hydroxyphenyl)diisopropylbenzenes together with the ring-brominated and/or ring-chlorinated derivatives thereof.

15 Particularly preferred diphenols are 4,4'-dihydroxydiphenyl, bisphenol A, 2,4-bis-(4-hydroxyphenyl)-2-methylbutane, 1,1-bis-(4-hydroxyphenyl)cyclohexane, 1,1-bis-(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane, 4,4'-dihydroxydiphenyl sulfide, 4,4'-dihydroxydiphenyl sulfone together with the di- and tetrabrominated or chlorinated derivatives thereof, such as for example 2,2-bis-(3-chloro-4-hydroxyphenyl)propane, 2,2-bis-(3,5-dichloro-4-hydroxyphenyl)propane or 2,2-bis-(3,5-dibromo-4-hydroxyphenyl)propane.

25 2,2-Bis-(4-hydroxyphenyl)propane (bisphenol A) is particularly preferred.

The diphenols may be used individually or as any desired mixtures.

30 The diphenols are known from the literature or are obtainable using processes known from the literature.

Chain terminators suitable for the production of the thermoplastic, aromatic polycarbonates are, for example, phenol, p-chlorophenol, p-tert.-butylphenol or 2,4,6-tribromophenol, as well as long-chain alkylphenols, such as 4-(1,3-tetramethylbutyl)phenol according to DE-OS 2 842 005 or monoalkylphenol or dialkylphenols having a total of 8 to 20 C atoms in the alkyl substituents, such as 3,5-di-tert.-butylphenol, p-iso-octylphenol, p-tert.-octylphenol, p-dodecylphenol and 2-(3,5-dimethylheptyl)phenol and 4-(3,5-dimethylheptyl)phenol. The quantity of chain terminators to be used is generally between 0.5 mol% and 10 mol%, relative to the sum of moles of the diphenols used in each case.

10

The thermoplastic, aromatic polycarbonates have weight average molecular weights (M_w , measured for example by ultracentrifugation or light scattering) of 10000 to 200000, preferably of 20000 to 80000.

15

The thermoplastic, aromatic polycarbonates may be branched in a known manner, preferably by incorporating 0.05 to 2.0 mol%, relative to the sum of diphenols used, of trifunctional or greater than trifunctional compounds, for example those having three and more than three phenolic groups.

20

Both homopolycarbonates and copolycarbonates are suitable. Component A copolycarbonates according to the invention may be produced by also using 1 to 25 wt.%, preferably 2.5 to 25 wt.% (relative to the total quantity of diphenols to be used) of polydiorganosiloxanes having hydroxy-aryloxy end groups. These are known (*c.f.* for example US patent 3 419 634) or may be produced using processes known from the literature. The production of copolycarbonates containing polydiorganosiloxanes is described, for example, in DE-OS 3 334 782.

25

30

Preferred polycarbonates, apart from bisphenol A homopolycarbonates, are copolycarbonates of bisphenol A with up to 15 mol%, relative to the sum of moles of diphenols, of other diphenols mentioned as preferred or particularly preferred, in particular 2,2-bis-(3,5-dibromo-4-hydroxyphenyl)propane.

Aromatic dicarboxylic acid dihalides for the production of aromatic polyester carbonates are preferably the diacid dichlorides of isophthalic acid, terephthalic acid, diphenyl ether 4,4'-dicarboxylic acid and 2,6-naphthalenedicarboxylic acid.

5

Mixtures of the diacid dichlorides of isophthalic acid and terephthalic acid in a ratio of between 1:20 and 20:1 are particularly preferred.

10

A carbonic acid halide, preferably phosgene, is additionally used as a difunctional acid derivative in the production of polyester carbonates.

15

Chain terminators which may be considered for the production of the aromatic polyester carbonates are, apart from the above-mentioned monophenols, also the chlorocarbonic acid esters thereof and the acid chlorides of aromatic monocarboxylic acids, which may optionally be substituted by C₁-C₂₂ alkyl groups or by halogen atoms, together with aliphatic C₂-C₂₂ monocarboxylic acid chlorides.

20

The quantity of chain terminators is in each case 0.1 to 10 mol%, relative, in the case of phenolic chain terminators, to the number of moles of diphenols and, in the case of monocarboxylic acid chloride chain terminators, to the number of moles of dicarboxylic acid dichlorides.

25

The aromatic polyester carbonates may also contain incorporated aromatic hydroxycarboxylic acids.

30

The aromatic polyester carbonates may be both linear and branched in a known manner (*c.f.* in this connection also DE-OS 2 940 024 and DE-OS 3 007 934).

Branching agents which may be used are, for example, tri- or polyfunctional carboxylic acid chlorides, such as trimesic acid trichloride, cyanuric acid trichloride, 3,3',4,4'-benzophenonetetracarboxylic acid tetrachloride, 1,4,5,8-naphthalene-

tetracarboxylic acid tetrachloride or pyromellitic acid tetrachloride, in quantities of 0.01 to 1.0 mol% (relative to dicarboxylic acid dichlorides used) or tri- or polyfunctional phenols, such as phloroglucinol, 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-2-heptene, 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)heptane, 1,3,5-tri-(4-hydroxyphenyl)benzene, 1,1,1-tri-(4-hydroxyphenyl)ethane, tri-(4-hydroxyphenyl)phenylmethane, 2,2-bis-[4,4-bis-(4-hydroxyphenyl)cyclohexyl]propane, 2,4-bis-(4-hydroxyphenylisopropyl)phenol, tetra-(4-hydroxyphenyl)methane, 2,6-bis-(2-hydroxy-5-methylbenzyl)-4-methylphenol, 2-(4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl)propane, tetra-(4-[4-hydroxyphenylisopropyl]phenoxy)methane, 1,4-bis[4,4'-dihydroxytriphenyl)methyl]benzene, in quantities of 0.01 to 1.0 mol%, relative to diphenols used. Phenolic branching agents may be introduced initially with the diphenols, acid chloride branching agents may be introduced together with the acid dichlorides.

The proportion of carbonate structural units in the thermoplastic, aromatic polyester carbonates may be varied at will. The proportion of carbonate groups is preferably up to 100 mol%, in particular up to 80 mol%, particularly preferably up to 50 mol%, relative to the sum of ester groups and carbonate groups. Both the ester and carbonate fractions of the aromatic polyester carbonates may be present in the form of blocks or randomly distributed in the polycondensation product.

The relative solution viscosity (η_{rel}) of the aromatic polycarbonates and polyester carbonates is in the range from 1.18 to 1.4, preferably from 1.22 to 1.3 (measured on solutions of 0.5 g of polycarbonate or polyester carbonate in 100 ml of methylene chloride solution at 25°C).

The thermoplastic, aromatic polycarbonates and polyester carbonates may be used alone or as any desired mixture with each other.

Component B

Component B comprises one or more graft polymers of

5 B.1 5 to 95, preferably 30 to 80 wt.%, of at least one vinyl monomer on

10 B.2 95 to 5, preferably 70 to 20 wt.%, of one or more grafting backbones having glass transition temperatures of $<10^{\circ}\text{C}$, preferably of $<0^{\circ}\text{C}$, particularly preferably of $<-20^{\circ}\text{C}$.

The grafting backbone B.2 generally has an average particle size (d_{50} value) of 0.05 to 5 μm , preferably of 0.10 to 0.5 μm , particularly preferably of 0.20 to 0.40 μm .

15 Monomers B.1 are preferably mixtures of

20 B.1.1 50 to 99 parts by weight of vinyl aromatics and/or ring-substituted vinyl aromatics (such as for example styrene, α -methylstyrene, p-methylstyrene, p-chlorostyrene) and/or (meth)acrylic acid (C_1 - C_8)-alkyl esters (such as for example methyl methacrylate, ethyl methacrylate) and

25 B.1.2 1 to 50 parts by weight of vinyl cyanides (unsaturated nitriles such as acrylonitrile and methacrylonitrile) and/or (meth)acrylic acid (C_1 - C_8)-alkyl esters (such as for example methyl methacrylate, n-butyl acrylate, t-butyl acrylate) and/or derivatives (such as anhydrides and imides) of unsaturated carboxylic acids (for example maleic anhydride and N-phenylmaleimide).

30 Preferred monomers B.1.1 are selected from among at least one of the monomers styrene, α -methylstyrene and methyl methacrylate, preferred monomers B.1.2 are selected from among at least one of the monomers acrylonitrile, maleic anhydride and methyl methacrylate.

Particularly preferred monomers are B.1.1 styrene and B.1.2 acrylonitrile.

5 Grafting backbones B.2 suitable for the graft polymers B are for example diene rubbers, EP(D)M rubbers, *i.e.* those based on ethylene/propylene and optionally diene, acrylate, polyurethane, silicone, chloroprene and ethylene/vinyl acetate rubbers.

10 Preferred grafting backbones B.2 are diene rubbers (for example based on butadiene, isoprene *etc.*) or mixtures of diene rubbers or copolymers of diene rubbers or mixtures thereof with further copolymerisable monomers (for example according to B.1.1 and B.1.2), preferably butadiene/styrene copolymers, providing that the glass transition temperature of component B.2 is $<10^{\circ}\text{C}$, preferably $<0^{\circ}\text{C}$, particularly preferably $<-10^{\circ}\text{C}$.

15 Pure polybutadiene rubber is particularly preferred.

20 Particularly preferred polymers B are, for example, ABS polymers (emulsion, bulk and suspension ABS), as are described for example in DE-OS 2 035 390 (= US patent 3 644 574) or in DE-OS 2 248 242 (= GB patent 1 409 275) or in Ullmann, *Enzyklopädie der Technischen Chemie*, volume 19 (1980), pp. 280 *et seq.*. The gel content of the grafting backbone B.2 is at least 30 wt.%, preferably at least 40 wt.% (measured in toluene).

25 The graft copolymers B are produced by free-radical polymerisation, for example by emulsion, suspension, solution or bulk polymerisation, preferably by emulsion polymerisation or bulk polymerisation.

30 Particularly suitable graft rubbers are ABS polymers which are produced by redox initiation using an initiator system comprising organic hydroperoxide and ascorbic acid according to US patent 4 937 285.

Since, as is known, the graft monomers are not necessarily grafted in their entirety onto the grafting backbone during the grafting reaction, graft polymers B are also taken according to the invention to include those products which are obtained by (co)polymerisation of the graft monomers in the presence of the grafting backbone and are also isolated during working up.

Suitable polymer B acrylate rubbers B.2 are preferably polymers prepared from acrylic acid alkyl esters, optionally with up to 40 wt.%, relative to B.2, of other polymerisable, ethylenically unsaturated monomers. Preferred polymerisable acrylic acid esters include C_1 - C_8 alkyl esters, for example methyl, ethyl, butyl, n-octyl and 2-ethylhexyl esters; haloalkyl esters, preferably halo- C_1 - C_8 -alkyl esters, such as chloroethyl acrylate and mixtures of these monomers.

Monomers having more than one polymerisable double bond may also be copolymerised for crosslinking purposes. Preferred examples of crosslinking monomers are esters of unsaturated monocarboxylic acids having 3 to 8 C atoms and unsaturated monohydric alcohols having 3 to 12 C atoms, or saturated polyols having 2 to 4 OH groups and 2 to 20 C atoms, such as for example ethylene glycol dimethacrylate, allyl methacrylate; polyunsaturated heterocyclic compounds, such as for example trivinyl and triallyl cyanurate; polyfunctional vinyl compounds, such as di- and trivinylbenzenes; as well as triallyl phosphate and diallyl phthalate.

Preferred crosslinking monomers are allyl methacrylate, ethylene glycol dimethacrylate, diallyl phthalate and heterocyclic compounds having at least three ethylenically unsaturated groups.

Particularly preferred crosslinking monomers are the cyclic monomers triallyl cyanurate, triallyl isocyanurate, triacryloylhexahydro-s-triazine, triallylbenzenes.

The quantity of crosslinking monomers is preferably 0.02 to 5, preferably 0.05 to 2 wt.%, relative to the grafting backbone B.2.

It is advantageous to limit the quantity of cyclic crosslinking monomers having at least three ethylenically unsaturated groups to below 1 wt.% of the grafting backbone B.2.

Preferred "other" polymerisable, ethylenically unsaturated monomers which, in addition to the acrylic acid esters, may optionally be used to produce the grafting backbone B.2 are, for example, acrylonitrile, styrene, α -methylstyrene, acrylamides, vinyl C₁-C₆-alkyl ethers, methyl methacrylate, butadiene. Preferred acrylate rubbers as the grafting backbone B.2 are emulsion polymers having a gel content of at least 60 wt.%.

Further suitable grafting backbones B.2 are silicone rubbers having active grafting sites, as are described in DE-OS 3 704 657, DE-OS 3 704 655, DE-OS 3 631 540 and DE-OS 3 631 539.

The gel content of the grafting backbone B.2 is determined in a suitable solvent at 25°C (M. Hoffmann, H. Krömer, R. Kuhn, *Polymeranalytik I & II*, Georg Thieme Verlag, Stuttgart 1977).

The average particle size d_{50} is the diameter both above and below which 50 wt.% of the particles lie. This value may be measured by ultracentrifugation (W. Scholtan, H. Lange, *Kolloid Z. und Z. Polymere*, 250 (1972), 782-1796).

Component C

Component C comprises one or more thermoplastic vinyl (co)polymers C.1. and/or polyalkylene terephthalates C.2.

Suitable vinyl (co)polymers C.1 are polymers of at least one monomer from the group of vinyl aromatics, vinyl cyanides (unsaturated nitriles), (meth)acrylic acid (C_1-C_8)-alkyl esters, unsaturated carboxylic acids as well as derivatives (such as anhydrides and imides) of unsaturated carboxylic acids. Particularly suitable (co)polymers are those prepared from

C.1.1 50 to 99, preferably 60 to 80 parts by weight of vinyl aromatics and/or ring-substituted vinyl aromatics (such as for example styrene, α -methylstyrene, p-methylstyrene, p-chlorostyrene) and/or (meth)acrylic acid (C_1-C_8)-alkyl esters (such as for example methyl methacrylate, ethyl methacrylate) and

C.1.2 1 to 50, preferably 20 to 40 parts by weight of vinyl cyanides (unsaturated nitriles) such as acrylonitrile and methacrylonitrile and/or (meth)acrylic acid (C_1-C_8)-alkyl esters (such as for example methyl methacrylate, n-butyl acrylate, t-butyl acrylate) and/or unsaturated carboxylic acids (such as maleic acid) and/or derivatives (such as anhydrides and imides) of unsaturated carboxylic acids (for example maleic anhydride and N-phenylmaleimide).

The (co)polymers C.1 are resinous, thermoplastic and rubber-free.

The copolymer of C.1.1 styrene and C.1.2 acrylonitrile is particularly preferred.

The (co)polymers C.1 are known and may be produced by free-radical polymerisation, in particular by emulsion, suspension, solution or bulk polymerisation. The (co)polymers preferably have molecular weights \overline{M}_w (weight average, determined by light scattering or sedimentation) of between 15000 and 200000.

The component C.2 polyalkylene terephthalates are reaction products of aromatic dicarboxylic acids or the reactive derivatives thereof, such as dimethyl esters or

anhydrides, and aliphatic, cycloaliphatic or araliphatic diols, together with mixtures of these reaction products.

5 Preferred polyalkylene terephthalates contain at least 80 wt.%, preferably at least 90 wt.%, relative to the dicarboxylic acid component, of terephthalic acid residues and at least 80 wt.%, preferably at least 90 mol%, relative to the diol component, of ethylene glycol and/or 1,4-butanediol residues.

10 In addition to terephthalic acid residues, the preferred polyalkylene terephthalates may contain up to 20 mol%, preferably up to 10 mol%, of residues of other aromatic or cycloaliphatic dicarboxylic acids having 8 to 14 C atoms or aliphatic dicarboxylic acids having 4 to 12 C atoms, such as for example residues of phthalic acid, isophthalic acid, 2,6-naphthalenedicarboxylic acid, 4,4'-diphenyldicarboxylic acid, succinic acid, adipic acid, sebacic acid, azelaic acid, cyclohexanediadicetic acid.

15 In addition to ethylene glycol or 1,4-butanediol residues, the preferred polyalkylene terephthalates may contain up to 20 mol%, preferably up to 10 mol%, of other aliphatic diols having 3 to 12 C atoms or cycloaliphatic diols having 6 to 21 C atoms, for example residues of 1,3-propanediol, 2-ethyl-1,3-propanediol, neopentyl glycol, 1,5-pentanediol, 1,6-hexanediol, 1,4-cyclohexanedimethanol, 3-ethyl-2,4-pentanediol, 2-methyl-2,4-pentanediol, 2,2,4-trimethyl-1,3-pentanediol, 2-ethyl-1,3-hexanediol, 2,2-diethyl-1,3-propanediol, 2,5-hexanediol, 1,4-di-(β -hydroxyethoxy)benzene, 2,2-bis-(4-hydroxycyclohexyl)propane, 2,4-dihydroxy-1,1,3,3-tetramethylcyclobutane, 2,2-bis-(4- β -hydroxyethoxyphenyl)propane and 2,2-bis-(4-hydroxypropoxyphenyl)propane (DE-OS 2 407 674, 2 407 776, 2 715 932).

30 The polyalkylene terephthalates may be branched by incorporating relatively small quantities of tri- or tetrahydric alcohols or tri- or tetrabasic carboxylic acids, for example according to DE-OS 1 900 270 and US-Patent 3 692 744. Examples of further preferred branching agents are trimesic acid, trimellitic acid, trimethylolethane and trimethylolpropane and pentaerythritol.

Particularly preferred polyalkylene terephthalates are those solely produced from terephthalic acid and the reactive derivatives thereof (for example the dialkyl esters thereof) and ethylene glycol and/or 1,4-butanediol, and mixtures of these polyalkylene terephthalates.

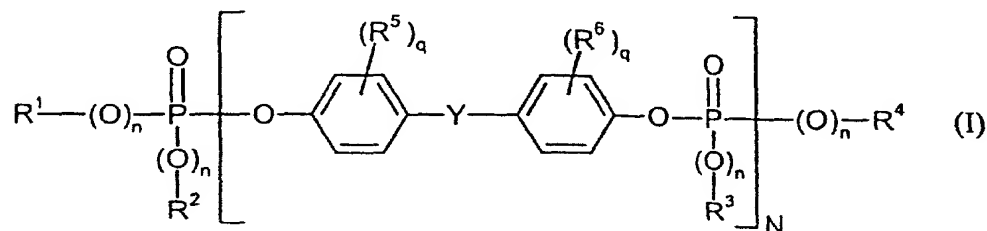
Mixtures of polyalkylene terephthalates contain 1 to 50 wt.%, preferably 1 to 30 wt.%, of polyethylene terephthalate and 50 to 99 wt.%, preferably 70 to 99 wt.%, of polybutylene terephthalate.

The preferably used polyalkylene terephthalates generally have an intrinsic viscosity of 0.4 to 1.5 dl/g, preferably of 0.5 to 1.2 dl/g, measured in phenol/o-dichlorobenzene (1:1 parts by weight) at 25°C in a Ubbelohde viscosimeter.

The polyalkylene terephthalates may be produced using known methods (*c.f.* for example *Kunststoff-Handbuch*, volume VIII, pp. 695 *et seq.*, Carl Hanser Verlag, Munich 1973).

Component D

The moulding compositions according to the invention contain phosphorus compounds according to formula (I) as flame retardants,



in which the residues have the above-stated meanings.

The component D phosphorus compounds which are suitable according to the invention are generally known (c.f. for example *Ullmanns Encyklopädie der technischen Chemie*, volume 18, pp. 301 *et seq.*, 1979; Houben-Weyl, *Methoden der organischen Chemie*, volume 12/1, page 43; Beilstein, volume 6, page 177).

5

Preferred substituents R^1 to R^4 comprise methyl, butyl, octyl, chloroethyl, 2-chloropropyl, 2,3-dibromopropyl, phenyl, cresyl, cumyl, naphthyl, chlorophenyl, bromophenyl, pentachlorophenyl and pentabromophenyl. Methyl, ethyl, butyl, phenyl and naphthyl are particularly preferred.

10

The aromatic groups R^1 , R^2 , R^3 and R^4 may be substituted with halogen and/or C_1 - C_4 alkyl. Particularly preferred aryl residues are cresyl, phenyl, xylenyl, propylphenyl or butylphenyl as well as the brominated and chlorinated derivatives thereof.

15

R^5 and R^6 mutually independently preferably mean methyl or bromine.

Y preferably denotes C_1 - C_7 alkylene, in particular isopropylidene or methylene.

n in the formula (I) may mutually independently be 0 or 1, n preferably equals 1.

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q may be 0, 1, 2, 3 or 4, q is preferably 0, 1 or 2, q particularly preferably equals 0.

N may assume values of 0.9 to 10, preferably of 0.95 to 5, in particular of 1 to 3. Mixtures of various phosphates may also be used as component D according to the invention. In this case, N has an average value. Monophosphorus compounds ($N = 0$) may also be present in this mixture.

25

The average N values may be determined by using suitable methods [gas chromatography (GC), high pressure liquid chromatography (HPLC), gas permeation chromatography (GPC)] to determine the composition of the phosphate

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mixture (molecular weight distribution) and calculating therefrom the average values for N.

Component E

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Fluorinated polyolefins may be added as a further component.

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The fluorinated polyolefins E are of a high molecular weight and have glass transition temperatures of above -30°C , generally of above 100°C , fluorine contents preferably of 65 to 76, in particular of 70 to 76 wt.%, average particle diameters d_{50} of 0.05 to 1000, preferably of 0.08 to 20 μm . The fluorinated polyolefins E generally have a density of 1.2 to 2.3 g/cm^3 . Preferred fluorinated polyolefins E are polytetrafluoroethylene, polyvinylidene fluoride, tetrafluoroethylene/hexafluoropropylene and ethylene/tetrafluoroethylene copolymers. The fluorinated polyolefins are known (c.f. *Vinyl & Related Polymers* by Schildknecht, John Wiley & Sons Inc., New York, 1962, pp. 484-494; *Fluoropolymers* by Wall, Wiley-Interscience, John Wiley & Sons Inc., New York, volume 13, 1970, pp. 623-654; *Modern Plastics Encyclopedia*, 1970-1971, volume 47, no. 10 A, October 1970, McGraw-Hill Inc., New York, pp. 134 and 774; *Modern Plastics Encyclopedia*, 1975-1976, October 1975, volume 52, no. 10A, McGraw-Hill Inc., New York, pp. 27, 28 and 472 and US-Patent 3 671 487, 3 723 373 and 3 838 092).

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They may be produced using known processes, thus for example by polymerising tetrafluoroethylene in an aqueous medium with a free-radical forming catalyst, for example sodium, potassium or ammonium peroxydisulfate, at pressures of 7 to 71 kg/cm^2 and at temperatures of 0 to 200°C , preferably at temperatures of 20 to 100°C . (c.f. for example US patent 2 393 967 for further details). Depending upon the form in which they are used, the density of these materials may be between 1.2 and 2.3 g/cm^3 , the average particle size between 0.5 and 1000 μm .

30

Fluorinated polyolefins E preferred according to the invention are used in the form of an emulsion having average particle diameters of 0.05 to 20 μm , preferably of 0.08 to 10 μm , and a density of 1.2 to 1.9 g/cm^3 , or in the form of powder having average particle diameters of 100 to 1000 μm and densities of 2.0 g/cm^3 to 2.3 g/cm^3 .

The fluorinated polyolefins E are used according to the invention in the form of special preparations:

- 10 E.1) as a coagulated mixture with at least one of components A to C, wherein the fluorinated polyolefin E or polyolefin mixture is mixed as an emulsion with at least one emulsion of components A to C and then coagulated,
or
- 15 E.2) as a precompound with at least one of components A to C, wherein the fluorinated polyolefins E are mixed as a powder with a powder or pellets of at least one of components A to C and melt-compounded, generally at temperatures of 200°C to 330°C, in conventional units such as internal kneaders, extruders or twin-screw extruders.

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Preferred preparations for the fluorinated polyolefins E are coagulated mixtures with a graft polymer B or a vinyl (co)polymer C.

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A coagulated mixture is produced from B and E by first mixing an aqueous emulsion (latex) of a graft polymer B with a finely divided emulsion of a tetrafluoroethylene polymer E; suitable tetrafluoroethylene polymer emulsions conventionally have solids contents of 30 to 70 wt.%, in particular of 50 to 60 wt.%, preferably of 30 to 35 wt.%.

The quantity of component A, B and C stated in the description does not include the proportion of the graft polymer, vinyl (co)polymer or polycarbonate for the coagulated mixture according to E.1 and E.2.

5 In the emulsion mixture, the equilibrium ratio of graft polymer B or (co)polymers to the fluorinated polyolefin E is 95:5 to 60:40, preferably 90:10 to 50:50. The emulsion mixture is then coagulated in a known manner, for example by spray drying, freeze drying or coagulation by addition of inorganic or organic salts, acids, bases or organic, water-miscible solvents, such as alcohols, ketones, preferably at
10 temperatures of 20 to 150°C, in particular of 50 to 100°C. If necessary, drying may be performed at 50 to 200°C, preferably at 70 to 100°C.

Suitable tetrafluoroethylene polymer emulsions are conventional commercial products and are offered for sale, for example, by DuPont (Wilmington, Delaware, USA) as Teflon® 30 N or by Dyneon GmbH (Burgkichen, Germany) as Hostaflon®.
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Preferred preparations for the fluorinated polyolefins E are furthermore precompounds with a polycarbonate A or a graft polymer B or a vinyl (co)polymer C.
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In the precompound, the ratio of component A, B or C to the fluorinated polyolefin E is 95:5 to 60:40, preferably 90:10 to 50:50.

25 Suitable fluorinated polyolefin powders are conventional commercial products and are offered for sale, for example, by DuPont as Teflon® CFP 6000 N or by Dyneon GmbH (Burgkichen, Germany) as Hostaflon® TF 2071.

The moulding compositions according to the invention may contain at least one of
30 the conventional additives, such as lubricants and mould release agents, for example

pentaerythritol tetrastearate, nucleating agents, anti-static agents, stabilisers, fillers and reinforcing materials as well as dyes and pigments.

5 The filled or reinforced moulding compositions may contain up to 60, preferably 10 to 40 wt.%, relative to the filled or reinforced moulding composition, of fillers and/or reinforcing materials. Preferred reinforcing materials are glass fibres. Preferred fillers, which may also have a reinforcing action, are glass beads, mica, silicates, quartz, talcum, titanium dioxide, wollastonite.

10 The moulding compositions according to the invention may contain up to 35 wt.%, relative to the overall moulding composition, of a further, optionally synergistic flame retardant. Examples of further flame retardants which may be mentioned are organic halogen compounds, such as decabromobisphenyl ether, tetrabromobisphenol, inorganic halogen compounds such as ammonium bromide,
15 nitrogen compounds, such as melamine, melamine/formaldehyde resins, inorganic hydroxide compounds, such as Mg, Al hydroxide, inorganic compounds such as antimony oxides, barium metaborate, hydroxoantimonate, zirconium oxide, zirconium hydroxide, molybdenum oxide, ammonium molybdate, zinc borate, ammonium borate, barium metaborate, talcum, silicate, silicon oxide and tin oxide,
20 as well as siloxane compounds. Monophosphate compounds, oligomeric phosphate compounds or mixtures thereof may additionally be used as flame retardants. Such phosphorus compounds are described in EP-A 363 608, EP-A 345 522 and DE-OS 197 21 628.

25 The moulding compositions according to the invention containing components A to E and optionally further known additives such as stabilisers, dyes, pigments, lubricants and mould release agents, nucleating agents, as well as antistatic agents, fillers and reinforcing materials are produced by mixing the particular constituents in a known manner and melt-compounding and melt-extruding them at temperatures of
30 200°C to 300°C in conventional units such as internal kneaders, extruders and twin-screw extruders.

The individual constituents may be mixed in a known manner both in succession and simultaneously and both at approx. 20°C (room temperature) and at a higher temperature.

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The present invention accordingly also provides a process for the production of the moulding compositions.

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The moulding compositions of the present invention may be used for the production of mouldings of any kind. Mouldings may in particular be produced by injection moulding. Examples of mouldings which may be produced are: casings of all kinds, for example for domestic appliances such as juice extractors, coffee machines, food mixers, for office equipment, such as monitors, printers, copiers or cladding sheet for the building sector and automotive components. They may also be used in

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electrical engineering applications as they have very good electrical properties.

The moulding compositions according to the invention may furthermore, for example, be used to produce the following mouldings or shaped articles:

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1. Interior trim for rail vehicles
2. Hub-caps
3. Casings for electrical devices containing small transformers
4. Casings for information dissemination and transmission devices
5. Casings and cladding for medical purposes

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6. Massage devices and casings therefor
7. Toy vehicles for children
8. Sheet wall elements
9. Casings for safety equipment
10. Hatchback spoilers

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11. Thermally insulated transport containers
12. Apparatus for keeping or caring for small animals

13. Mouldings for sanitary and bathroom installations
14. Cover grilles for ventilation openings
15. Mouldings for summer houses and sheds
16. Casings for garden appliances.

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Another processing method is the production of mouldings by thermoforming of previously produced sheet or film.

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The present invention accordingly also provides the use of the moulding compositions according to the invention for the production of mouldings of all kinds, preferably those stated above, and the mouldings made from the moulding compositions according to the invention.

Examples

Component A

- 5 Bisphenol A based polycarbonate having a relative solution viscosity of 1.278, measured in methylene chloride at 25°C and a concentration of 0.5 g/100 ml.

Component B

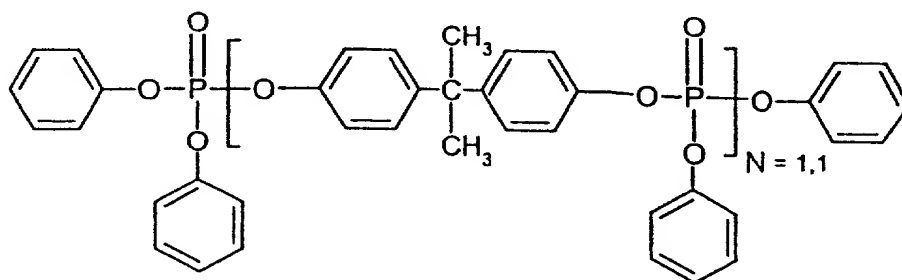
- 10 Graft polymer of 40 parts by weight of styrene and acrylonitrile in a 73:27 ratio on 60 parts by weight of particulate crosslinked polybutadiene rubber (average particle diameter $d_{50} = 0.28 \mu\text{m}$), produced by emulsion polymerisation.

Component C

- 15 Styrene/acrylonitrile copolymer having a styrene:acrylonitrile ratio of 72:28 and an intrinsic viscosity of 0.55 dl/g (measured in dimethylformamide at 20°C).

Component D

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25

The average N value was determined by firstly determining the proportions of the monomeric and oligomeric phosphates by HPLC measurements:

30

Column type: LiChrosorp RP-8
Eluent in the gradient: Acetonitrile/ water 50:50 to 100:0
Concentration: 5 mg/ml

- 5 The number-weighted averages are then determined in known manner from the proportions of the individual constituents (mono- and oligophosphates).

Component E

10 E.1 (Comparison)

PTFE powder Teflon CFP 6000 N (DuPont, Wilmington, Delaware, USA)

E.2

- 15 Coagulated mixtures prepared from 90 parts by weight of graft polymer (of 40 parts by weight of styrene and acrylonitrile in a ratio of 73:27 on 60 parts by weight of particulate crosslinked polybutadiene rubber having an average particle diameter $d_{50} = 0.28 \mu\text{m}$) and 10 parts by weight of polytetrafluoroethylene polymer (Teflon® 30 N).

20 E.3

Coagulated mixture prepared from 80 parts by weight of graft polymer (as E.2) and 20 parts by weight of polytetrafluoroethylene polymer (Teflon® 30 N).

E.4

- 25 Coagulated mixture prepared from 50 parts by weight of vinyl (co)polymer (of styrene and acrylonitrile in a ratio of 72:28, intrinsic viscosity of 0.85 dl/g measured in dimethylformamide at 20°C) and 50 parts by weight of polytetrafluoroethylene polymer (Teflon® 30 N).

E.5

Precompound prepared from 80 parts by weight of graft polymer (of 84 parts by weight of styrene and acrylonitrile in a ratio of 72:28 on 16 parts by weight of crosslinked butadiene rubber) and 20 parts by weight of polytetrafluoroethylene polymer (Teflon® CFP 6000 N).

E.6

Precompound prepared from 90 parts by weight of vinyl (co)polymer (of styrene and acrylonitrile in a ratio of 72:28, intrinsic viscosity of 0.55 dl/g measured in dimethylformamide at 20°C) and 10 parts by weight of polytetrafluoroethylene polymer (Teflon® CFP 6000 N).

E.7

Precompound prepared from 90 parts by weight of polycarbonate (based on bisphenol A having a relative solution viscosity of 1.278, measured in methylene chloride at 25°C and a concentration of 0.5 g/100 ml) and 10 parts by weight of polytetrafluoroethylene polymer (Teflon® CFP 6000 N).

Production and testing of the moulding compositions according to the invention

The components are mixed with conventional processing auxiliaries in a ZSK 32 twin screw extruder. The mouldings are produced at 260°C on an Arburg model 270 E injection moulding machine.

Notched impact strength is determined in accordance with method ISO 180 IA on bars of dimensions $80 \times 10 \times 4 \text{ mm}^3$ at room temperature.

The Vicat B softening point is determined according to DIN 53 460 on bars of dimensions $80 \times 10 \times 4 \text{ mm}^3$.

Elongation at break is determined according to ISO 527/DIN 53 457.

Flame retardancy is determined according to UL 94V.

Stress cracking behaviour (ESC behaviour) is investigated on bars of dimensions $80 \times 10 \times 4 \text{ mm}^3$, injection moulding temperature 260°C. The test medium used is a mixture of 60 vol.% toluene and 40 vol.% isopropanol. The test pieces are pre-stressed on a circular arc template (initial elongation ϵ_x in percent) and immersed in the test medium at room temperature. Stress cracking behaviour is evaluated on the basis of cracking or failure as a function of initial elongation in the test medium.

Table 1**Composition and properties of moulding compositions**

	1 (Comp.)	2	3	4	5	6	7
Components [parts by weight]							
A	68.4	68.4	68.4	68.4	68.4	68.4	64.8
B	9.4	5.8	7.8	9.4	8.9	9.0	9.4
C	9.3	9.3	9.3	8.9	8.1	5.7	9.3
D	12.5	1.25	12.5	12.5	12.5	12.5	12.5
E.1	0.4						
E.2	-	4.0	-	-	-		-
E.3	-	-	2.0	-	-		-
E.4	-	-	-	0.8	-		-
E.5	-	-	-	-	2.0		-
E.6	-	-	-	-	-	4.0	-
E.7	-	-	-		-		4.0
Properties:							
Vicat B 120 [°C]	102	103	103	103	103	102	103
a_k ISO 180 1A [kJ/m ²]	42	47	50	48	48	46	45
Elongation at break [%]	35	56	58	65	75	70	68
ESC behaviour, failure at ϵ_x [%]	1.8	2.0	2.4	2.4	2.4	2.0	2.0
UL 94 V at 1.6 mm							
Rating	V-2	V-0	V-0	V-0	V-0	V-0	V-0
Total burn time [sec]	47	29	27	22	19	28	26

- 5 The moulding compositions according to the invention, *i.e.* those which were produced from special PTFE preparations, are distinguished by distinctly improved mechanical properties (notched impact strength (a_k), elongation at break), greater

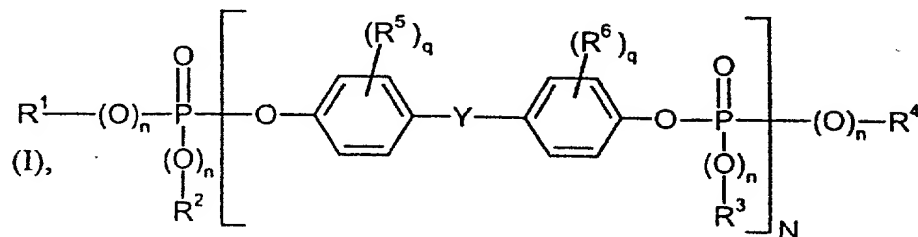
stress cracking resistance as well as improved flame retardancy in accordance with the UL 94 V test (reduction of formation of burning droplets, reduced burn time). Having this range of properties, the moulding compositions according to the invention are in particular suitable for flame resistant housing components of a complicated geometry which are mechanically stressed and exposed to materials.

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Patent Claims

1. Graft polymer-modified polycarbonate moulding compositions containing phosphorus compounds of the formula (I)



in which

R^1 , R^2 , R^3 and R^4 are mutually independently C_1 - C_8 alkyl, optionally substituted by halogen, C_5 - C_6 cycloalkyl, C_6 - C_{10} aryl or C_7 - C_{12} aralkyl, each optionally substituted by halogen and/or alkyl,

n are mutually independently 0 or 1,

q are mutually independently 0, 1, 2, 3 or 4,

N is 0, 9 to 10,

R^5 and R^6 mutually independently mean C_1 - C_4 alkyl or halogen,

Y means C_1 - C_7 alkylidene, C_1 - C_7 alkylene, C_5 - C_{12} cycloalkylene, C_5 - C_{12} cycloalkylidene, -O-, -S-, -SO-, -SO₂- or -CO-,

and fluorinated polyolefins in the form of a coagulated mixture or as a precompound.

2. Moulding compositions according to claim 1, containing the fluorinated polyolefin or polyolefin mixture as

1) a coagulated mixture with at least one component selected from among graft polymer, copolymer and polycarbonate, wherein the fluorinated polyolefins are mixed as an emulsion with at least one emulsion of the stated components and then coagulated,

or

2) as a precompound with at least one component selected from among graft polymer, copolymer and polycarbonate, wherein the fluorinated polyolefins are mixed as a powder with a powder or pellets of at least one of the stated components and melt-compounded.

3. Moulding compositions according to claim 1, containing 0.5 to 20 parts by weight of phosphorus compound of the formula (I) or mixtures thereof.

4. Moulding compositions according to claims 1 to 3, containing 0.01 to 3 parts by weight of fluorinated polyolefin in the form of a coagulated mixture or as a precompound.

5. Moulding compositions according to claim 4, containing 0.05 to 2 parts by weight of fluorinated polyolefin.

6. Moulding compositions according to claim 1, containing

A) 40 to 99 parts by weight of aromatic polycarbonate and/or polyester carbonate

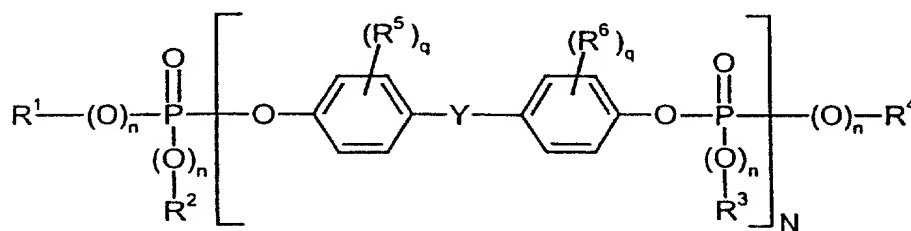
B) 0.5 to 60 parts by weight of graft polymer of

B.1) 5 to 95 wt.% of one or more vinyl monomers on

B.2) 95 to 5 wt.% of one or more grafting backbones having a glass transition temperature of $<10^{\circ}\text{C}$,

C) 0 to 45 parts by weight of at least one thermoplastic polymer selected from the group comprising vinyl (co)polymers and polyalkylene terephthalates,

D) 0.5 to 20 parts by weight of a phosphorus compound of the formula (I)



in which R^1 to R^6 , Y, n, N and q have the meaning stated in claim 1,

E) 0.01 to 3 parts by weight of fluorinated polyolefins according to claim 1.

7. Moulding compositions according to one of the preceding claims, wherein N in the formula (I) denotes an average value of 0.95 to 5.

8. Moulding compositions according to one of the preceding claims, wherein N in the formula (I) denotes an average value of 1 to 3.

9. Moulding compositions according to one of the preceding claims, containing fluorinated polyolefin or polyolefin mixture

1) as a coagulated mixture with at least one of components A to C, wherein the fluorinated polyolefin E or polyolefin mixture is mixed as an emulsion with at least one emulsion of components A to C and then coagulated,

or

2) as a precompound with at least one of components A to C, wherein the fluorinated polyolefins E are mixed as a powder with a powder or pellets of at least one of components A to C and melt-compounded.

10. Moulding compositions according to one of the preceding claims containing one or more graft polymers of

B.1 5 to 95 wt.% of at least one vinyl monomer on

B.2 95 to 5 wt.% of one or more grafting backbones having glass transition temperatures of $< 10^{\circ}\text{C}$.

11. Moulding compositions according to claim 10, containing as the vinyl monomers B.1 mixtures of

B.1.1 50 to 99 parts by weight of vinyl aromatic and/or ring-substituted vinyl aromatics and/or (meth)acrylic acid ($\text{C}_1\text{-C}_8$)-alkyl esters and

B.1.2 1 to 50 parts by weight of vinyl cyanides and/or (meth)acrylic acid ($\text{C}_1\text{-C}_8$)-alkyl esters and/or derivatives of unsaturated carboxylic acids.

12. Moulding compositions according to one of the preceding claims, containing a diene rubber, acrylate rubber, silicone rubber or ethylene/propylene/diene rubber or mixtures thereof as the grafting backbone B.2.

13. Moulding compositions according to one of the preceding claims, wherein Y in the formula (I) denotes isopropylidene or methylene.

14. Moulding compositions according to claim 13, wherein Y in the formula (I) denotes isopropylidene.

15. Moulding compositions according to one of the preceding claims, containing vinyl (co)polymers of at least one monomer from the group comprising vinyl aromatics, vinyl cyanides, (meth)acrylic acid (C₁-C₈)-alkyl esters, unsaturated carboxylic acids, as well as derivatives (such as anhydrides and imides) of unsaturated carboxylic acids.

16. Moulding compositions according to one of the preceding claims, containing at least one addition from the group comprising stabilisers, pigments, mould release agents, flow auxiliaries and/or antistatic agents, fillers and reinforcing materials.

17. Use of the moulding compositions according to one of the preceding claims for the production of mouldings.

18. Mouldings obtainable from moulding compositions according to one of the preceding claims.

Flame resistant, graft polymer-modified polycarbonate moulding compositions

A b s t r a c t

This invention relates to flame resistant graft polymer-modified polycarbonate moulding compositions containing phosphorus compounds and specially prepared fluorinated polyolefins, which compositions exhibit excellent flame retardancy and very good mechanical properties such as notched impact strength, elongation at break and very good stress cracking behaviour.

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

FLAME-RESISTANT POLYCARBONATE MOULDING MATERIALS MODIFIED WITH GRAFT POLYMERS

the specification of which is attached hereto,

or was filed on **March 14, 2000**

as a PCT Application Serial No. **PCT/EP00/02242**

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s), the priority(ies) of which is/are to be claimed:

199 14 137.1
(Number)

Germany
(Country)

March 27, 1999
(Month/Day/Year Filed)

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose the material information as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)	(Filing Date)	(Status)
		(patented, pending, abandoned)

(Application Serial No.)	(Filing Date)	(Status)
		(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Le A 33 592-US

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